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Impact of flaxseed gum on the aggregate structure, pasting properties, and rheological behavior of waxy rice starch

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ABSTRACT

This study examines the effects of flaxseed gum (FG) on the aggregate structure, pasting and rheological properties of waxy rice starch (WRS). Results display an increase in the ordered molecular structure ($R_{1047/1024}$), relative crystallinity (*RC*), compactness (α), and microphase heterogeneity (ε, density degree of nanoaggregates, from 3.52 to 4.23) for WRS-FG complexes. These suggested FG facilitated the development of more organized molecular and crystalline structures of WRS, accompanied by the formation of ordered nanoaggregates with higher density (*i.e.*, nano-aggregation structure). Also, FG addition resulted in the formation of enhanced gel network structure characterized by thicker layer walls and more uniform pores. These structural transformations contributed to a rise in gelatinization temperature (T_o, from 56.90 °C to 62.10 °C) and enthalpy (ΔH), as well as alterations in paste viscosities (PV, from 1285.00 mPa·s to 1734.00 mPa·s), and the rigidity of network structure (*e.g.*, decreased loss tangent). These results indicate that FG could effectively regulate the techno-functional properties of WRS by rationally controlling the starch intrinsic structures of starch. And this study may improve the pasting and gelling properties of starch, thus driving the development of high-quality starchy foods and prolonging their shelf life, especially for glutinous rice flour products.

1. Introduction

Waxy rice, a prominent rice variety cultivated in Asia, particularly in China, possesses distinct characteristics, including high viscosity, tender texture, and excellent transparency [1,2]. It has been widely applied to produce diverse foods such as traditional Chinese products, sauces, baby foods and puddings. However, food products prepared by raw waxy rice or waxy rice flour (WRF) have disadvantages such as weak gel structure, high digestibility and limited characteristic viscosity, which seriously affected the development of rice products and consumers' acceptability. And these deficiencies may be determined by the technological properties of WRF, especially for thickening and gelling properties [3]. In other words, enhancing the favorable swelling ability, pasting properties, and rheological behavior of waxy rice or waxy rice flour can address these quality shortcomings. Therefore, in order to develop overall quality of final products, there is considerable attention focused on how to regulate the thickening and gelling behaviors of WRF.

As the main constituent in waxy rice, starch (*ca.* 85 %, dry basis) plays an important role in determining the thickening and gelling behaviors of waxy rice or WRF, eventually affecting the quality attributes of related products [4]. That is to say, the main technological properties influenced are basically the thickening and gelling behaviors that are indicated by starch viscosity and gelling parameters, respectively. The modified characteristics of starch may change the sensory properties and make the end product preferable. Thus, it is valuable for designing products with the desired quality by rationally controlling the pasting

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and rheological behaviors of starch. Currently, numerous studies showed that the combination of starch and hydrocolloids had gained an increasing attention due to that these complexes or matrices exhibited good gelling and thickening ability and strong enzymatic resistance [5–7]. Thereinto, xanthan gum (XG), guar gum (GG), konjac glucomannan (KGM) and tamarind seed gum (TSG) are common edible gums (*i.e.*, hydrocolloids) that utilized for addressing technological deficiencies such as low shape retention and thickening ability, as well as weak gel structure in starch or starch-based foods [8–10]. For example, previous studies had demonstrated that these hydrocolloids could significantly increase characteristic viscosity, gel intensity and the fraction of resistant starch through intermolecular steric interaction or intra- and inter-chain interactions/entanglements (*i.e.*, starch-hydrocolloid interactions) between starch and hydrocolloids [11,12]. Nevertheless, reversed results had been found in other studies, and these phenomena may be ascribed to the differences in structural characteristics of hydrocolloids and starches [8]. These contradictory results observed in different studies might be attributed to variations in starch or hydrocolloid structural characteristics such as molecular flexibility, molecular chain length, aggregate structures, charges and constituent of heteropolysaccharide [13,14]. Thus, it can be proposed that effects of hydrocolloids on starch pasting and rheological behaviors are strongly influenced by the type of hydrocolloid, the origin of starch, as well as the preparation conditions and concentrations.

Flaxseed gum (FG) is a natural polysaccharide extracted from flaxseeds. It is comprised by a mixture of polysaccharides including galactose, arabinose, glucose, xylose, rhamnose, fucose, and galacturonic acid, which displays distinct techno-functional properties such as good water retention, emulsification, stability, thickening and gel properties [6,15]. Flaxseed flour had been extensively used to enhance the elasticity and palatability of food products due to its good gelling, thickening and emulsification ability [16]. For example, Marpalle et al. [17] had reported that flaxseed flour could improve the water absorption and stickiness of dough as well as crumb softness of bread, which is more prominent with increased flaxseed level. Research by Papagianni et al. [18] also revealed that the addition of flaxseed flour to the gluten-free dough reduced crumb hardness while increasing specific loaf volume. Despite the extensive use of flaxseed flour in food technology, there is scarce information about the impact of their hydrocolloid nature and the interactions between starch polymer chains when they are dissolved or dispersed. To the best of our knowledge, some studies suggested that FG addition could modulate the pasting and retrogradation properties of starch [19,20]. But how FG addition affected the aggregate structure (helical structure, short-range ordered molecular structure, crystalline structure and nano-aggregation structure), pasting properties, and rheological behavior of waxy rice starch has received little attention. Furthermore, it is not yet able to perfectly model or predict the physicochemical properties and structural changes of starch-hydrocolloid systems through previous studies because of their complexity. Therefore, information obtained in this work will aid in further understanding the interaction between starch and hydrocolloids, and this combination may improve the stability and texture of host foods, and they may also retard the retrogradation of many starch-based products. And this work may expand the development of high-quality starchy foods (*e.g.*, glutinous dumplings and puddings) and increase the economic effect of FG and waxy rice starch (WRS) in the food industry.

The study aimed to clarify the interaction between WRS and FG, with a specific focus on understanding how FG addition affected the aggregate structure and physicochemical properties of WRS. Specifically, this work focused on assessing the alterations of helical, short-range ordered, crystalline, lamellar and granular structure; as well as the thermal and pasting properties, rheological behavior and digestibility of waxy rice starch with the presence of FG. These transformations were conducted using the following techniques: rapid visco analyzer (RVA), differential scanning calorimetry (DSC), rotational rheometer, nuclear magnetic resonance spectrometer (^{13}C CP/MAS NMR), Fourier infrared spectroscopy (FT-IR), X-ray diffraction (XRD), small-angle X-ray scattering (SAXS) and scanning electron microscopy (SEM) [21]. And our results may promote the application of hydrocolloid in developing starch-based food products with good quality, especially for improving the stability and texture of host foods. Furthermore, it has never been more important to understand the relationship among starch-hydrocolloid interactions, functionality and quality attributes of related products added with hydrocolloids, which is valuable for elevating the overall quality of final products in the future.

2. Materials and methods

2.1. Materials

Waxy rice starch (WRS) was kindly supplied by Huangguo Grains Industry Co., Ltd. (Xinyang, Henan, China), and its molecular weight and moisture content were 2.20 × 10^7 g/mol and 13.2 % (*w*/w), respectively. And its apparent viscosity at room temperature (*ca.* 25 °C) is no >50 mPa·s. Flaxseed gum (FG, 6.7 % moisture content, 1 % viscosity at 25 °C was 326 mPa·s) was obtained from Xinjiang Linseed Biologic Technologies Co., Ltd. (Yili, Xinjiang, China). Amyloglucosidase (cat. A3306, 318 U/mL) and *α*-amylase (cat. P-7545, activity 8 × USP/g) were purchased from Sigma-Aldrich (St. Louis, MO, USA), and the glucose oxidase/peroxidase (GOPOD) assay kit was provided by Megazyme Co., Ltd. (Wicklow, Ireland). All other chemical reagents used were of analytical grade, which were purchased from Sinopharm Chemical Reagent Co., Ltd. (Shanghai, China).

2.2. Starch isolation

Starch was extracted from waxy rice flour according to the procedure outlined by Liu et al. [1]. Initially, about 100 g of waxy rice flour was immersed in 0.2 % sodium hydroxide solution (500 mL) with continuously stirring (300 rpm) for 4.0 h at 25 °C. The slurries were neutralized with 1 M HCl to pH 7 and stored at 4 °C for 24 h. Then, the supernatant was removed and the precipitate was re-dispersed with 500 mL of deionized water, followed by the centrifugation at 4000 r/min for 15 min at room temperature (*ca.* 25 °C). This procedure was repeated twice. Afterwards, the purified starch was subsequently dried at 45 °C in a drying oven for 24 h, then passed through a 100 μm screen, and carefully sealed.

2.3. Preparation of WRS/FG samples

FG was added to distilled water at varying ratios of 0 %, 0.1 %, 0.3 %, 0.5 %, 0.7 %, and 0.9 % (*w*/w, dry basis) and magnetically stirred (500 rpm) at 30 °C for 4 h. Subsequently, WRS (6 %, w/w, dry basis) was added to different FG solutions for obtaining starch-FG complexes. Specifically, sample preparation was conducted by heating starch-FG mixtures at 95 °C for 30 min and then cooling to 25 °C. Lastly, the frozen starch samples (freezing at −80 °C) were dried with a freeze dryer (12 N/A, Ningbo Scientz Biotechnology Co., LTD, China), subsequently crushed and filtered through a 160-mesh sieve. The nomenclature assigned to the WRS/FG complexes reflected their FG concentrations, labeled as "WRS/0.1%FG", "WRS/0.3%FG", "WRS/0.5%FG", "WRS/0.7%FG", and "WRS/0.9%FG", where the numerical values represent the specific concentrations of FG added.

2.4. Thermal properties

Thermal behaviors of all samples were conducted using a differential scanning calorimeter (DSC Q2000, TA Instrument, Newcastle, USA). Each sample was precisely weighed (3 mg, dry basis) into an aluminum pan (Tzero® pan, TA), then added with 7 μL deionized water and hermetically sealed. After an equilibration period of 24 h at 4 °C, the prepared mixtures were heated from 30 °C to 100 °C at 10 °C/min heating

rate, whilst an empty aluminum pan was used as the control. The results provided information on the enthalpy of pasting (ΔH) and the pasting transition temperature, including the onset temperature (T_o), peak temperature (T_p), and conclusion temperature (T_c).

2.5. Pasting properties

Pasting properties were estimated using a Rapid Visco Analyzer (RVA 4800, Perten Instruments, Sweden). The WRS and WRS/FG samples (6 %, *w*/w, dry basis) were stirred at 35 °C for 1 min, then heated to 95 °C and equilibrated at 95 °C for 10 min, followed by cooling to 50 °C and maintained at 50 °C for 10 min [22]. The heating and cooling rates were both set to 5 °C/min, and the stirring speed was 160 rpm, except for the initial 60 s (960 rpm). The pasting temperature (PT), peak viscosity (PV), breakdown viscosity (BD), and setback viscosity (SB) were calculated based on the analysis.

2.6. Rheological properties

The rheological properties of WRS and WRS/FG mixtures were evaluated using an HR-1 rotational rheometer (TA Instruments, New Castle, USA). Starch gel obtained from RVA was placed on a 40 mm plate, and all samples were edge-trimmed to ensure proper testing (1.05 mm). Silicone oil was applied to prevent moisture evaporation during testing. A test gap of 1 mm and the oscillation frequency mode were chosen. A strain of 1 % was ultimately selected. The frequency range within 0.1–100 rad/s was recorded (30 °C), and values of storage modulus (G'), loss modulus (G''), and loss factor (tan δ) were summarized.

2.7. In vitro digestibility measurement

The digestion properties of WRS and WRS/FG mixtures were assessed following a modified protocol based on prior research [23]. The mixed enzyme solution was prepared as follows: 12 g of porcine pancreatic α-amylase was dispersed in 80 mL of deionized water with magnetical stirring and centrifuged at 3000 *g* for 25 min. The supernatant (54 mL) was transferred into a beaker and mixed with 2.8 mL of amyloglucosidase and 3.2 mL of deionized water. The mixed enzyme solution was freshly prepared before use.

Lyophilized WRS and WRS/FG samples were dispersed in a 20 mL sodium acetate buffer solution (0.1 mol/L, pH 5.2). Subsequently, a mixture of α-amylase and amyloglucosidase solutions (5 mL) was added and hydrolysed at 37 °C. At intervals of 20 and 120 min, 0.5 mL of the hydrolysate was obtained and then deactivated using a 70 % ethanol solution (20 mL). The resultant mixtures underwent centrifugation at 4500 rpm for 20 min, and 0.1 mL of the supernatant was extracted to measure glucose content utilizing a GOPOD kit. Glucose levels at different hydrolysis durations (20 and 120 min) were recorded, allowing for the quantification of rapidly digestible starch (RDS), slowly digestible starch (SDS), and resistant starch (RS).

2.8. Helical structure measurement

Helical structure analysis was performed using a ^{13}C CP/MAS nuclear magnetic resonance spectrometer (^{13}C CP/MAS NMR, Advance III HD 400, Bruker Inc., Germany) at a resonance frequency of 100.6 MHz, which was equipped with a 4-mm probe (double-resonance MAS), followed by the methodology of previous research by Tan et al. [24] with minor modifications. Lyophilized WRS and WRS/FG samples (500 mg) were packed into a 4 mm ZrO_2 rotor and spun at the magic angle (54.7°) with 6 kHz of spin. Each spectrum was recorded over 2400 scans with a constant contact time (2.5 ms) and recycle delay (2 s). The raw data were recorded and analyzed with Peak-Fit software (version 4.1, America) to calculate the contents of double helices, single helices, and amorphous material.

2.9. Short-range ordered structure measurement

The short-range ordered structure was evaluated using a Fourier-transform infrared spectrometer (FTIR, Vertex 70, Bruker Inc., Germany). The lyophilized WRS or WRS/FG samples were mixed with KBr at a ratio of 1:200 (*w*/w, dry basis) and compressed into tablets. The scanning range was 4000 to 400 cm^{-1} at a scanning resolution of 4 cm^{-1} [25,26]. Spectra within the range of 1200–800 cm^{-1} were automatically baseline corrected, normalized, and deconvoluted with a half band width of 19 cm^{-1} and an enhancement factor of 1.9. The $R_{1047/1022}$ ratio of the deconvoluted spectra was determined for each sample by measuring the amplitudes at bands located at 1047 cm^{-1} and 1022 cm^{-1}.

2.10. Crystalline structure measurement

The crystalline structure was evaluated using an X-ray diffractometer (XRD, mod D8, Bruker Inc., Karlsruhe, Germany) with Cu-Kα (λ = 0.1542 nm) radiation. The lyophilized WRS or WRS/FG samples were scanned over the 2θ range of 5°- 40° with a step size of 0.02°. The voltage was set as 40 kV, and the current was 40 mA. Before analysis, all wheat starches were conditioned to a constant humidity of about 11.0 % using saturated lithium chloride solution in a sealed container. The starch relative crystallinity (*RC*) was calculated by evaluating the area of crystalline peaks to the total diffraction area, using MDI Jade 6.0 software.

2.11. Lamellar structure measurement

The lamellar structure was measured using a small-angle X-ray scattering facility (SAXS, Rigaku S-MAX3000, Rigaku Co., Ltd., Tokyo, Japan), following the methodology of previous research by Wang et al. [27]. The freeze-dried WRS and WRS/FG samples were exposed to an X-ray monochromatic beam for 5 min at 40 kV and 50 mA, with an incident monochromatic light wavelength λ = 0.15 Å. All obtained data were normalized, background-subtracted, and processed through SAXS quant software. To gain a better understanding of the changes in nano-aggregated structures, the fractal dimension and structure density of the gel samples were evaluated.

For a more in-depth analysis of the lamellar structure of starch, the Kratky scattering curve ($I(q)*q^2 \sim q$) was derived through Lorentz transformation. The scattering peak observed in the Kratky curve serves as a determinant of the degree of microphase heterogeneity within polymers. Additionally, the Debye-Bueche (DB) Eq. (1) was employed to fit samples exhibiting structural inhomogeneity:

$$I_{(q)} = \frac{I_{DB(0)}}{(1+\varepsilon^2 q^2)^2} + Aq^{-\delta} \quad (1)$$

where $\frac{I_{DB(0)}}{(1+\varepsilon^2 q^2)^2}$ represents the Debye-Bueche equation; ε denotes the characteristic length of starch inhomogeneity degree, nm; $Aq^{-\delta}$ is the Power-Law function, and A and δ are the prefactor and exponent of the Power-Law function, respectively.

2.12. Microstructure

Microstructure analysis was conducted using a JSM-7001F scanning electron microscope (SEM, Japan Electronics Co., Tokyo, Japan). Samples of WRS or WRS/FG after freeze-drying (a cross section) were affixed onto conductive tape and coated with gold. Microstructural observation was conducted at a 1.0 kV voltage and 300 × magnification.

2.13. Statistical analysis

All measurements were conducted in triplicates, and the results were presented as means with standard deviations. Differences between the

means of each individual group were determined (*i.e.*, One-way analysis of variance (ANOVA)) using Duncan's multiple range test with version 21.0 of IBM SPSS Statistics (IBM Corp, Armonk, NY, USA). A value of $P < 0.05$ was determined to indicate a statistically significant difference between two samples throughout the study using the SPSS software. All tests were implemented at least in triplicate.

3. Results and discussion

3.1. Thermal properties

Fig. 1a and Table 1 present the DSC curves and thermal parameters for WRS and WRS/FG mixtures. When compared to WRS, WRS/FG mixtures exhibited higher T_o and T_c, indicating higher thermostability. With the growing amount of FG, the T_o value gradually increased from 56.9 °C to 62.1 °C and the T_c value from 90.4 °C to 92.5 °C. This may be attributed to hydrophilic gum competing for available water during gelatinization, which decreased the water accessibility for starch molecules and thus elevated the starch melting temperature. Also, the potential interaction between FG and leached amylose/amylopectin may be responsible for the higher gelatinization temperatures in WRS/FG mixtures [20]. Xie et al. [28] similarly noted that interactions between starch and hydrocolloids enhanced T_o and T_c in corn starch.

Gelatinization enthalpy (ΔH) signifies the energy required for depolymerizing ordered molecular structures (double-helical and crystalline structures) [6]. The addition of FG led to an increase in ΔH, especially with higher amounts. This may be due to FG interacting with leached amylopectin chains and damaged starch molecules during gelatinization, reinforcing the formation of a polymeric network through hydrogen bonds. This results in a densely packed microscopic structure, consequently improving the ordered degree of molecular structures. Varela et al. [29] also reported that the addition of arabic gum increased the value of conclusion temperature (T_c) and enthalpy (ΔH) of the starch-hydrocolloid mixture, which may be due to the enhanced interconnections between hydrocolloid and water molecules (*i.e.*, the hydroxyl groups of FG and water molecules), thus reducing the available water absorbed by starch molecules during gelatinization, eventually interfering the gelatinization process and increasing the conclusion temperature (T_c) and gelatinization enthalpy of starches with FG addition.

3.2. Pasting properties

Starch gelatinization involves water absorption, amylose leaching, and amylopectin swelling [30]. Pasting curves for WRS and WRS/FG mixtures are displayed in Fig. S1, with corresponding viscosity parameters summarized in Table 1. Fig. S1 illustrates that peak viscosity (PV), final viscosity (FV), breakdown (BD), and setback (SB) of WRS/FG mixtures increased with higher FG concentrations. In Table 1, WRS exhibited the lowest PV and FV values of 1285.00 mPa·s and 839.00 mPa·s, respectively, rising to 1734.00 mPa·s and 1238.00 mPa·s upon the addition of 0.9 % (*w*/w) flaxseed gum. The enhanced characteristic viscosity (*i.e.*, increased PV and FV) might be interpreted as a result of a combination of phase separations, *i.e.*, the flaxseed gum could be excluded from gradually swollen starch granules during heating, which caused an increase in the effective concentration of hydrocolloids located in the continuous phase, subsequently resulting in high viscosity of a mixed system [31]. On the other hand, the interactions between

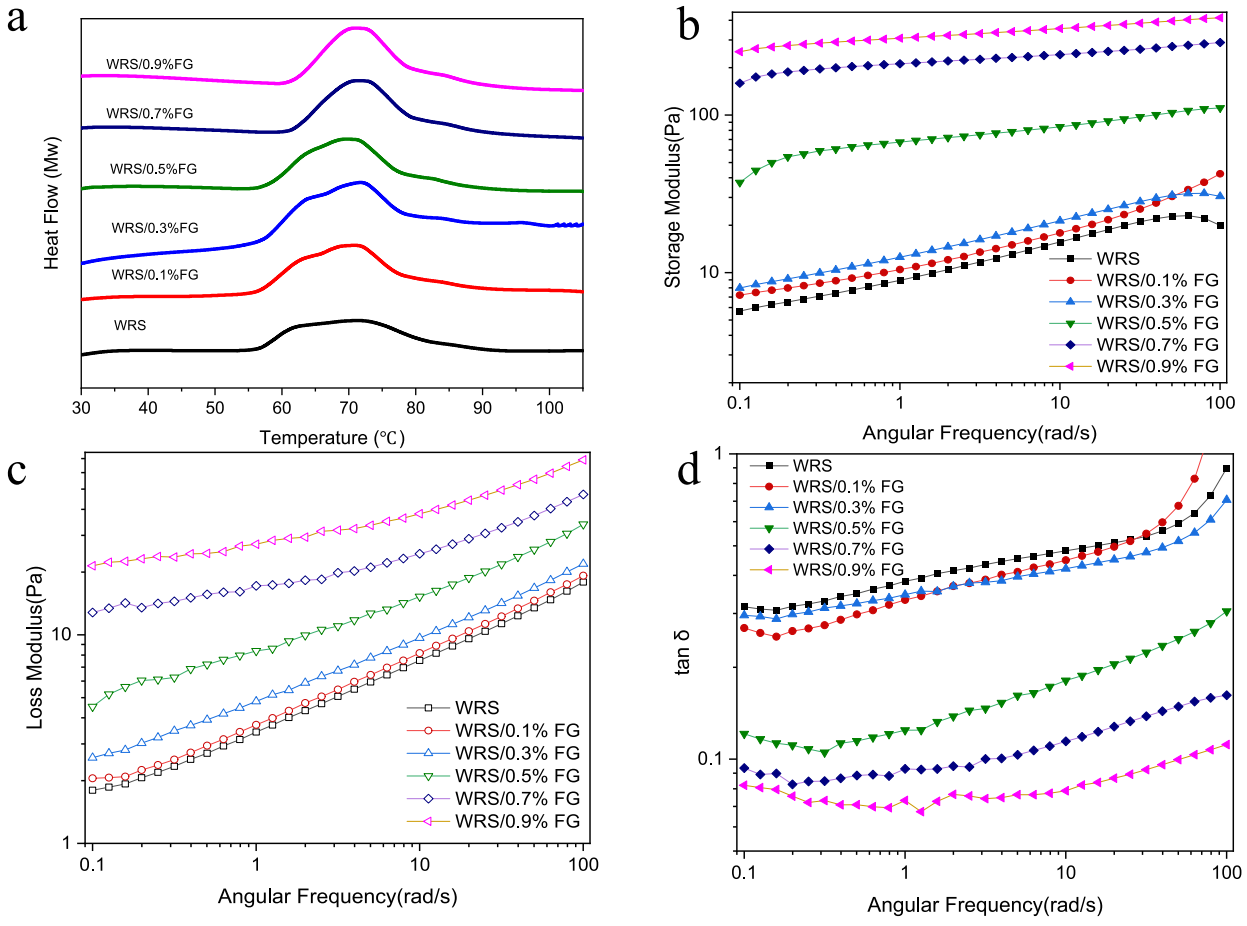

Fig. 1. DSC thermograms (a) and rheological behavior (b, c, d) of waxy rice starch (WRS) with the addition of flaxseed gum (FG). (b) Storage modulus, G'; (c) loss modulus, G''; (d) loss factor, tan δ.

Table 1
Effect of FG addition on the thermal properties, pasting properties and *in vitro* digestibility of waxy rice starch (WRS).[A,B]

Sample	WRS	WRS/0.1%FG	WRS/0.3%FG	WRS/0.5%FG	WRS/0.7%FG	WRS/0.9%FG
T_o (°C)	56.90 ± 0.40^{d}	56.60 ± 0.30^{d}	57.40 ± 0.50^{c}	58.20 ± 0.30^{b}	61.90 ± 0.20^{a}	62.10 ± 0.40^{a}
T_p (°C)	71.40 ± 0.80^{a}	70.70 ± 0.90^{a}	71.50 ± 0.70^{a}	69.40 ± 1.10^{a}	71.80 ± 0.50^{a}	71.20 ± 0.70^{a}
T_c (°C)	90.40 ± 1.20^{c}	90.70 ± 1.40^{c}	91.80 ± 1.00^{ab}	91.40 ± 1.30^{b}	92.40 ± 0.90^{a}	92.50 ± 1.10^{a}
ΔH (J/g)	10.70 ± 0.50^{d}	11.60 ± 0.60^{c}	12.20 ± 0.70^{c}	13.50 ± 0.50^{b}	14.10 ± 0.80^{a}	14.30 ± 0.70^{a}
PT (°C)	59.58 ± 0.58^{a}	60.38 ± 0.18^{a}	60.70 ± 0.28^{a}	60.38 ± 0.18^{a}	60.08 ± 0.25^{a}	60.40 ± 0.71^{a}
PV (mPa·s)	1285.00 ± 24.27^{f}	1322.00 ± 4.24^{e}	1379.00 ± 2.83^{d}	1493.50 ± 2.12^{c}	1629.00 ± 5.66^{b}	1734.00 ± 5.66^{a}
TV (mPa·s)	678.33 ± 21.13^{f}	714.50 ± 4.95^{e}	765.50 ± 0.71^{d}	811.00 ± 4.24^{c}	862.50 ± 12.02^{b}	905.50 ± 9.19^{a}
FV (mPa·s)	839.00 ± 19.29^{f}	899.50 ± 0.71^{e}	994.50 ± 3.54^{d}	1076.50 ± 10.61^{c}	1159.00 ± 7.07^{b}	1238.00 ± 7.07^{a}
BD (mPa·s)	606.67 ± 3.21^{d}	607.50 ± 0.71^{d}	613.50 ± 2.12^{d}	682.50 ± 6.36^{c}	766.50 ± 6.36^{b}	828.50 ± 14.85^{a}
SB (mPa·s)	160.67 ± 4.93^{f}	185.00 ± 5.66^{e}	229.00 ± 2.83^{d}	265.50 ± 6.36^{c}	296.50 ± 4.95^{b}	332.50 ± 2.12^{a}
RDS (%)	54.97 ± 1.16^{a}	52.48 ± 2.74^{ab}	50.11 ± 1.36^{bc}	48.00 ± 1.90^{cd}	47.18 ± 1.25^{d}	44.32 ± 1.43^{e}
SDS (%)	16.86 ± 1.72^{c}	17.49 ± 1.72^{bc}	20.12 ± 0.73^{ab}	20.46 ± 1.52^{ab}	21.16 ± 2.41^{a}	21.13 ± 0.89^{a}
RS (%)	28.16 ± 1.12^{d}	29.66 ± 1.06^{cd}	30.51 ± 0.61^{bc}	31.54 ± 1.48^{b}	32.03 ± 1.27^{b}	34.55 ± 0.55^{a}

[A] Reported values correspond to the mean ± standard deviation. T_o, onset temperature; T_p, peak temperature; T_c, conclusion temperature; ΔH, gelatinization enthalpy. PT, pasting temperature; PV, peak viscosity; FV, final viscosity; BD, breakdown; SB, setback value; RDS, rapidly digestible starch; SDS, slowly digestible starch; RS, resistant starch.

[B] Means with different superscript letters in the same line represent significant differences at $P < 0.05$.

starch molecular chains and polysaccharide residues of FG were considered to be an important reason for the viscosity increases [32]. That is to say, the interaction between starch and the hydrophilic gum could promote the hydration and fragmentation of starch granules and chain association, *i.e.*, facilitating the formation of hydrogen bonds, molecular entanglement and aggregation, oriented arrangement and packing of polymeric chains in this composite system, which resulted in an increased swelling power of starch granules as well as the robustness and rigidity of swollen granules, thus elevating PV and FV of WRS.

Heating and mechanical shearing are vital processes in food handling such as sterilization, cooling and storage [33]. The BD aims to reflect the stability of the granules against mechanical shear and heat upon continuous cooking [34]. In this work, BD showed an upward trend from 606.67 mPa·s to 828.50 mPa·s, suggesting that the addition of FG rendered the mixture less tolerant to the thermal and mechanical forces. In the case of starch-hydrocolloid paste, an increase in viscosity makes the shear forces excreted on the swollen granules much larger than those encountered in starch-water suspensions, thus resulting in higher breakdown values [35]. Liu et al. [6] also demonstrated that the shear stress on swollen granules with high viscosity was significantly greater, potentially disrupting starch granule integrity and leading to increased BD value. Furthermore, a similar trend was observed in SB, suggesting that FG addition facilitated short-term retrogradation. Xiao et al. [36] found that the phase separation between gum and starch promoted the rearrangement of starch molecular chains, thus enhancing the SB value. This could be due to the increase in the effective concentration of amylose in the continuous phase by adding gums, generating a substantial increase in final viscosity. On the other hand, the interactions between starch molecular chains and gum molecules may be partially responsible for the increase of setback value [37].

3.3. Rheological properties

Generally, the processing suitability and gelling properties of the food products or composite hydrogel materials can be predicted according to transformations of dynamic rheological behaviors, especially for the frequency sweep [38]. Fig. 1b–d shows the correlation between FG addition and variations in storage modulus (G'), loss modulus (G''), and loss factor (tan δ) with frequency in the composite system. As shown in Fig. 1b and c, the storage modulus (G') was significantly higher than the loss modulus (G'') in WRS gel, and both G' and G'' were increased with the increase of angular frequency, indicating that a solid-like behavior (a typical weak gel) was formed. Similar with the RVA results, FG affected the viscoelasticity remarkably. Both G' and G'' were increased with FG addition in an FG concentration-dependent manner, suggesting that FG could interact with WRS to form a compact three-dimensional network structure. A denser three-dimensional structure formed in the WRS/FG matrix due to the association between starch-starch and starch-hydrocolloid molecules [39], which are supposed to be largely correlated with their molecular flexibility and a persistence length of approximately 100 nm, as well as fractions containing greater amounts of acidic polysaccharides [40]. In addition, the presence of flaxseed gum may significantly increase the effective concentration of starch and decreased water activity, which promoted the association between starch molecules, given that chain-chain association competes with chain-water association [35].

Regarding tan δ, at the same angular frequency, the addition of flaxseed gum induced a decrease in magnitude, particularly noticeable for FG amounts exceeding 0.3 % (Fig. 1d). This implies that FG addition promoted the transition from a weak gel to a strong gel. The WRS/FG gel formed a more elastic and rigid network structure. It seems possible that FG facilitated the molecular entanglement and interaction between hydrocolloids and starch molecules, leading to the formation of rearranged molecular structures and the development of a sturdy gel. Also, Zheng et al. [41] discovered that hydrocolloids could be embedded into the three-dimensional network of starch, creating a relatively stable, rigid structure.

3.4. In vitro digestibility

Table 1 reveals that the RDS, SDS, and RS contents of WRS were 54.97 %, 16.86 %, and 28.16 %, respectively. The introduction of FG led to the conversion of some RDS into SDS and RS. As FG addition increased from 0.1 % to 0.9 %, RDS gradually decreased from 52.48 % to 44.32 %, and RS increased from 29.66 % to 34.55 %. Besides, these changes exhibited a positive correlation with the rise in FG concentration, thus slowing the digestion rate and extent of starch gel. The decrease in digestibility may be due to the increased viscosity of starch-hydrocolloid system, which impeded diffusion and adsorption of amylolytic enzymes to substrates [42]. Moreover, hydrophilic colloid might promote the interactions between hydrophilic polymer and starch molecular chains, which formed stable and ordered structures, hence leading to the reduction of starch digestibility. Previous studies also revealed that starch fortified with hydrocolloids tended to possess higher viscosity and/or a more ordered hierarchical structure, which could retard starch digestibility [43–45].

3.5. Helical structure

The solid-state NMR spectra of WRS and WRS/FG gels are shown in Fig. S2, and the detailed parameters of helical structures (double helices, single helix, and amorphous proportion) calculated are outlined in

Table 2
Effect of FG addition on the helical structure, short-range ordered molecular structure, crystalline structure, and nano-aggregated structure of waxy rice starch (WRS).[A,B]

Sample	WRS	WRS/ 0.1% FG	WRS/ 0.3% FG	WRS/ 0.5%FG	WRS/ 0.7%FG	WRS/ 0.9% FG
Double helix (%)	11.20 ± 0.50^{a}	9.70 ± 0.60^{b}	8.20 ± 0.30^{c}	7.90 ± 0.40^{cd}	7.70 ± 0.50^{d}	6.60 ± 0.30^{e}
Amorphous (%)	88.80 ± 1.60^{d}	90.30 ± 1.40^{c}	91.80 ± 2.10^{b}	92.10 ± 2.30^{ab}	92.30 ± 1.70^{ab}	93.40 ± 1.50^{a}
$R_{1047/1024}$	0.82 ± 0.02^{d}	0.83 ± 0.02^{cd}	0.85 ± 0.03^{c}	0.86 ± 0.01^{c}	0.89 ± 0.01^{b}	0.92 ± 0.02^{ab}
RC (%)	4.00 ± 0.20^{a}	4.40 ± 0.10^{a}	5.10 ± 0.30^{b}	5.50 ± 0.20^{b}	5.70 ± 0.10^{b}	6.00 ± 0.20^{b}
α	2.27 ± 0.02^{e}	2.32 ± 0.01^{d}	2.40 ± 0.04^{c}	2.45 ± 0.03^{c}	2.49 ± 0.02^{b}	2.54 ± 0.04^{a}
D_m	2.27 ± 0.02^{e}	2.32 ± 0.01^{d}	2.40 ± 0.04^{c}	2.45 ± 0.03^{c}	2.49 ± 0.02^{b}	2.54 ± 0.04^{a}
ε	3.52 ± 0.04^{f}	3.74 ± 0.02^{e}	3.88 ± 0.05^{d}	4.03 ± 0.03^{c}	4.18 ± 0.04^{b}	4.23 ± 0.06^{a}
R^2	0.999	0.997	0.999	0.999	0.998	0.999

[A] $R_{1047/1024}$, the ratio of peak intensity at 1047 cm^{-1} to 1024 cm^{-1}; *RC*, relative crystallinity; α, fractal dimension; D_m, mass fractal; ε, characteristic length.
[B] Means with different superscript letters in the same line represent significant differences at $P < 0.05$.

Table 2. Liu et al. [1] found that triple peaks in the C1 signal region for native starch indicated the A-type crystalline polymorph. However, in this current study, the starch gel exhibited no triple peaks at 100–104 ppm (Fig. S2), signifying that gelatinization disrupted the crystalline structure of WRS. Table 2 reveals that the WRS gel comprised 88.8 % amorphous and 11.2 % double-helix contents. The thermal energy from heating and the plasticizing effect of water molecules may disrupt both intermolecular and intramolecular hydrogen bonds within starch granules, leading to a reduction in the proportion of double-helix structures [46].

With the addition of FG, the amorphous content slightly increased from 88.8 % to 93.4 %, while the double-helical fragment decreased from 11.2 % to 6.6 % in a concentration-dependent manner. This observation suggests that FG inclusion potentially impeded the rearrangement or realignment of starch molecular chains, consequently diminishing the presence of the double-helical fraction. Notably, the abundance of hydrophilic groups in FG promoted the formation of intermolecular hydrogen bonds with starch molecular chains, thereby restraining molecular chain rearrangement or realignment (*e.g.*, amylose and amylopectin) and reducing the proportion of the double-helical fragment [20].

3.6. Short-range ordered structure

Fig. 2a presents the FT-IR spectra of WRS and WRS/FG gels. Compared with the WRS sample, no new peaks emerged after complexing with FG, implying that FG addition did not alter the chemical

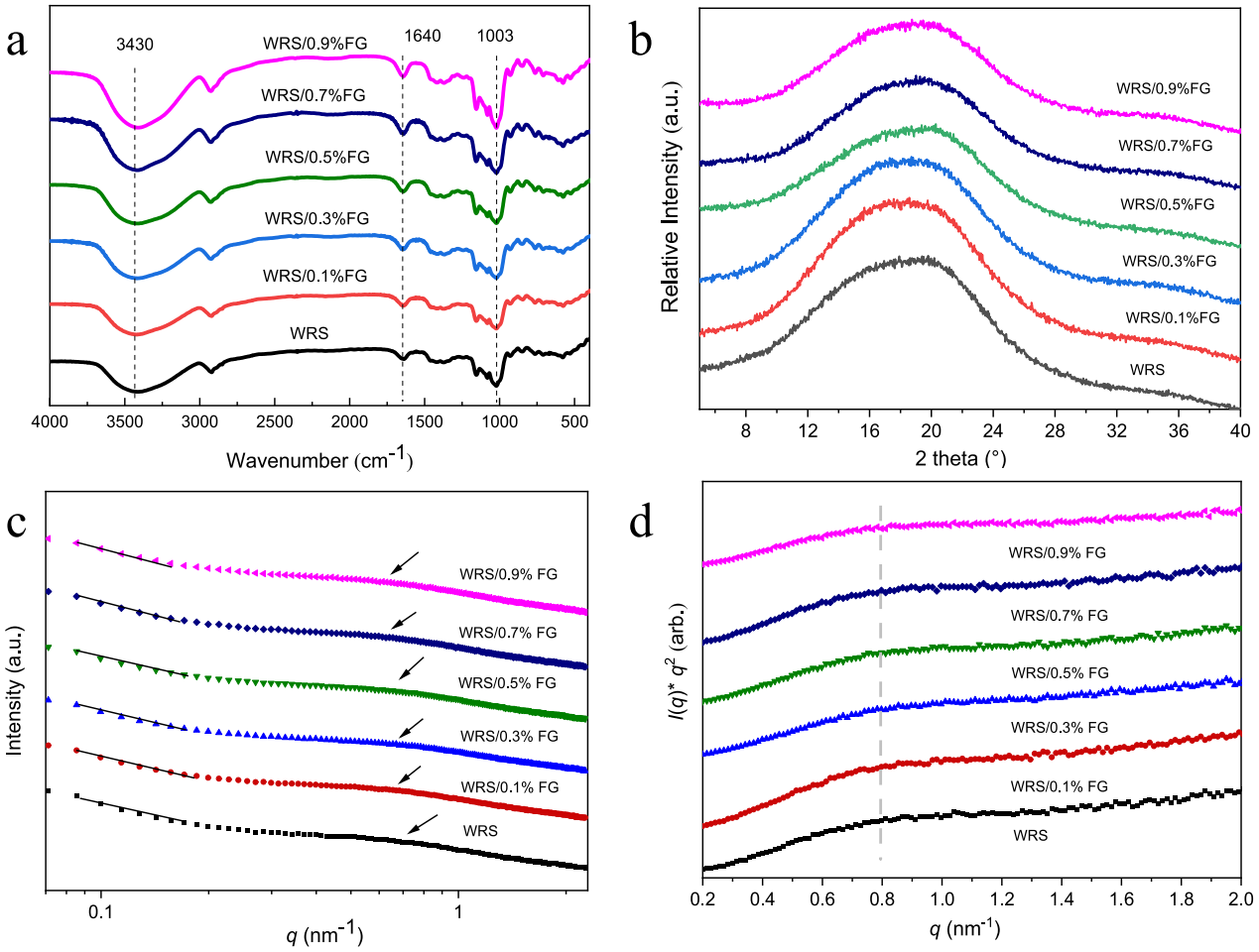

Fig. 2. FTIR spectra (a), X-ray diffraction patterns (b), double-logarithmic SAXS patterns (c), and Kratky curves (d) of waxy rice starch (WRS) with the addition of flaxseed gum (FG).

structure of WRS. Typically, absorption peaks at 3700–3000 cm^{-1} and 1640 cm^{-1} are primarily associated with -OH stretching and bending vibrations, representing intermolecular hydrogen bonds, while the absorption peak at 1000 cm^{-1} relates to intramolecular hydrogen bonding. From Fig. 2a, the peak intensity of WRS/FG gels at 3430 cm^{-1}, 1640 cm^{-1}, and 1000 cm^{-1} bands increased with the addition of FG, gradually rising with increasing FG amount. This suggests that FG might contribute to the formation of hydrogen bonds in the WRS/FG gels, aligning with the findings of a previous study by Wang et al. [22].

The absorption peaks at 1047 and 1024 cm^{-1} are correlated with crystalline and amorphous structures, respectively, and the $R_{1047/1024}$ value can reveal information about short-range ordered molecular structures [21]. As depicted in Fig. S3 and Table 2, the addition of FG improved the $R_{1047/1024}$ value of WRS/FG gels, ranging from 0.82 to 0.92. With the increase in FG concentration, the $R_{1047/1024}$ value gradually increased, implying the enhancement of the short-range ordered structure. This observation is consistent with DSC result. It is proposed that FG affected the formation of hydrogen bonds with amylose and amylopectin through interactions and entanglement between FG and starch molecules, which enhanced a stable and ordered molecular structure. In other words, the potential interactions and entanglement between hydrophilic polymer and amylopectin/damaged starch fragments may result in the formation of intermolecular or intramolecular hydrogen bonds for WRS-hydrocolloid matrix, thus forming a more stable and ordered molecular structure [13,47].

3.7. Crystalline structure

The XRD profiles of WRS and WRS/FG gels are displayed in Fig. 2b. According to Fig. 2b, WRS exhibited an A-type crystalline structure with diffraction peaks at 15°, 17°, 18°, and 23° (2θ). However, the characteristic diffraction peaks of all WRS/FG gels vanished, indicating the destruction of the crystalline structure of starch after gelatinization. It has been established that starch molecular chains rearrange after heating and cooling, resulting in the appearance of single or narrow peaks in starch gel [48]. Interestingly, WRS/FG gels exhibited diffused and broad peaks between 12° and 24° (2θ), possibly due to interactions between starch and FG, leading to the formation of a long-range ordered structure. From Table 2, the relative crystallinity (*RC*) of the WRS was 4.0 %, while the *RC* of all WRS/FG gels significantly increased, ranging from 4.4 % to 6.0 % with the rise in FG concentrations. The augmented *RC* might be attributed to the fact that FG facilitated the entanglement and interaction between polysaccharide chains and starch molecules during the heating process, *i.e.*, forming intermolecular hydrogen bonds with starch molecular chains, which facilitated the packing, arrangement or stacking of ordered structure. Ultimately, WRS/FG gels formed a newly-packed molecular structure, generating an ordered crystalline structure. The alteration of crystalline structure is in accordance with DSC and FTIR results.

3.8. Lamellar structure

SAXS is a valuable technique for investigating and characterizing the submicroscopic structure of polymer gels. The double-logarithmic patterns of gel samples are presented in Fig. 2c. An evident peak in the SAXS curves (*ca.* 0.65 nm^{-1}) reflected the information about the starch lamellar structure, such as the integrity and ordered degree of lamellar structure [49]. However, as depicted in Fig. 2c, a nearly disappeared peak was observed for all starch gels, with no new scattering peaks appearing. The result implies that the original lamellar structure of native WRS was disrupted after heating, leading to the disorganization of the lamellar structure.

Furthermore, the fractal structures of WRS and WRS/FG gel samples were calculated using the power-law fitting function ($I \sim q^{-\alpha}$). When the α value is between 1 and 4, it is mainly related to the accumulation of nano-scale structures (*e.g.*, ordered packing, arrangement, or stacking) in starch materials. A greater α value indicates denser structures of the scattering objects. For α values ranging from 1 to 3, the scattering object is categorized as a "mass fractal structure" with a fractal dimension of D_m, while a surface fractal structure is described with a fractal dimension ($D_s = 6 - \alpha$) when the α value falls between 3 and 4 [27]. The α value was calculated by the slope in the low q region of SAXS curves (Fig. 2c). Table 2 shows that the α values of all samples fall in the range of 1 to 3, indicating WRS and WRS/FG gels all had mass fractal structures. In comparison with WRS, the α value of WRS/FG gels showed a noticeable increasing trend with increasing FG concentration. This phenomenon demonstrates that FG enhanced the density or compactness of aggregated structures in WRS/FG gels, promoting the formation of a more densely packed nano-aggregation structure. It is likely that hydrophilic groups in FG readily interacted with starch molecular chains to form intermolecular hydrogen bonds, facilitating the stacking, alignment (*e.g.*, short-range ordered and crystalline structures), resulting in denser aggregated structures and improved starch densities at the mesoscale [50].

The Kratky scattering curves (Fig. 2d), obtained by Lorentz transformation, along with characteristic length of inhomogeneities (ε) and coefficients of determination (R^2), are presented in Table 2. The experimental data fit well with the Debye-Bueche (DB) equation (R^2 values higher than 0.99). The ε value of WRS gel was 3.52 nm, while FG addition significantly increased the ε value (from 3.52 to 4.23 nm) with rising FG concentration, indicating an increase in the inhomogeneity of WRS/FG gel. During the heating process of starch, ordered molecular and supramolecular structures were disrupted, causing separation, unfolding, or irregular curled and resulting in a lower ε value for WRS. Moreover, FG may promote rearrangement and reaggregation between the hydrocolloid and starch molecular chains, leading to the formation of a new ordered nano-aggregation structure. The new nano-aggregated structure likely enhanced the microphasic heterogeneity of WRS/FG gels, becoming more pronounced with an increase in FG amount. Consequently, it can be concluded that FG enhanced the ordered degree, density, and rigidity of the nano-aggregated structure of starch gel.

3.9. Microstructure

Fig. 3 reveals micrographs of all WRS/FG gels, showcasing three-dimensional porous structures with no remaining swollen granules. This indicates the complete gelatinization of WRS during heating, forming honeycomb-like porous structures akin to maize starch gels [7]. The honeycomb-like porous structures in the starch gel may result from water separation during freeze-drying, with pore size representing the magnitude of ice crystals [35]. In this context, the pore size of the WRS gel displayed an irregular feature with thin walls. Upon the addition of FG, the structure of the WRS/FG gel underwent significant changes. The WRS/FG gel displayed a compact network structure with uniform and large pores, presenting an enhanced skeleton structure. These changes become more pronounced with the increase in FG concentration. This transformation could be ascribed to the heightened interactions and entanglement between FG and leached amylopectin chains or damaged starch fragments (inferred from Sections 3.6–3.8), leading to the formation of an ordered and rigid honeycomb-structure structure. This phenomenon indicates that the FG enhanced the mechanical stability of WRS/FG gels. This may be ascribed to the enhanced ordered molecular structure, crystalline structure, and lamellar structure of WRS/FG gel, thus leading to increased starch enzymatic resistance. Moreover, the denser and more rigid network structure may also diminish the digestibility of starch.

3.10. Overall discussion

Starch-hydrocolloid combinations have been used in processed foods (including confections) for many years. Addition of the proper hydrocolloid can also overcome the shortcomings of native starches, for

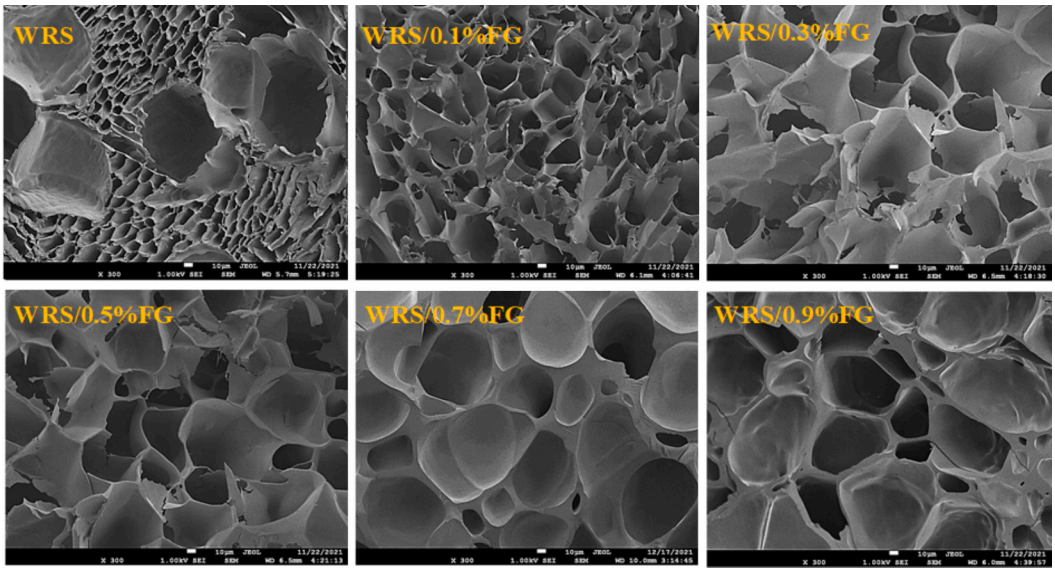

Fig. 3. SEM photographs of waxy rice starch (WRS) with the addition of flaxseed gum (FG).

example, can protect starch granules against shear during cooking, improve product texture/rheology, hold moisture, and protect against syneresis [13]. Specifically, the main properties influenced are basically the pasting and rheological properties that are indicated by viscosity and mechanical modulus, respectively. Thus, the processing suitability and gelling properties of the starch-hydrocolloid products can be predicted according to transformations of pasting and rheological behaviors. In order to disclose how FG addition affects the pasting and rheological properties of waxy rice starch, structural evolutions of starch with or without FG were investigated (shown in Fig. 4). Then, understanding the relationship between the structural features of starch-hydrocolloid complexes and their pasting rheological behaviors could provide a theoretical basis on rationally controlling the quality attributes of final products.

The peak viscosity (PV) reflects the resistance ability of starch to shearing during the heating stage before physical breakdown. Because of continuous swell of starch granules, peak viscosity can be acquired from a complete rupture of its inherent hierarchical structures [13]. Generally, structural disorganizations at the molecular and supramolecular levels could result in the weakened entanglements and interactions among molecular chains (*i.e.*, amylose and amylopectin) and fragile granules, which can decrease the swelling extent of starch granules [1]. In this work, FG addition may facilitate the interactions and entanglement between hydrophilic polymer and amylopectin/damaged starch fragments, which may form a more stable and ordered molecular structure (Sections 3.1 and 3.6), crystalline structure (Section 3.7), nano-aggregation structure (Section 3.8) and enhance the density, compactness and microphase heterogeneity (*i.e.*, increased ε) of starch-hydrocolloid complexes, thus increasing the PV of starch. Furthermore, starch-FG complexes presented tighter network structure with more uniform and larger pores, and the thickness of skeleton structure (Section 3.9) also increased after FG addition, these alterations also could improve the viscosity. Besides, the setback (*i.e.*, short-term retrogradation) increased with the increase of FG concentration, presumably due to the increase in the effective concentration of amylose in the continuous phase by adding gums, generating a substantial increase in final viscosity. On the other hand, the interactions between starch molecular chains and gum molecules may be partially responsible for the increase of setback value [37].

Following the gelling of starch composite system (*i.e.*, starch-flaxseed gum), different gel rigidity (G' and tan δ) exhibited in our research. During the gelling process, the inter-molecular and intra-molecular interactions among starch molecular chains (*i.e.*, amylose and amylopectin) and FG (*e.g.*, galactose, arabinose, glucose, and xylose) facilitated the rearrangement and/or realign of nano-aggregates, as evidenced by the increased *RC*, short-range ordered degree and density,

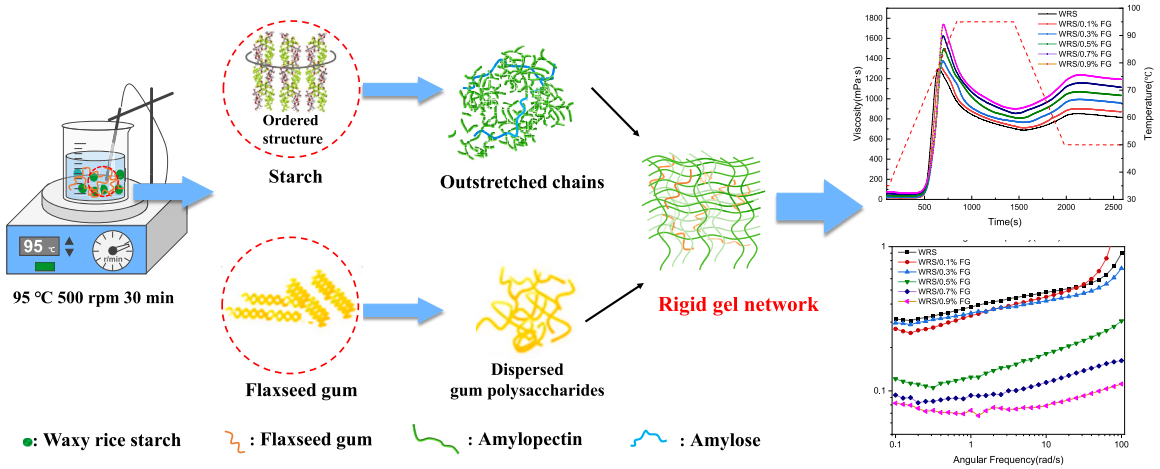

Fig. 4. Schematic showing how FG addition affects the aggregation structure, pasting and rheological behaviors of waxy rice starch.

as well as the compactness inhomogeneity of nano-aggregated structures. These aggregate structures contributed to enhancing the ordered degree, density, and rigidity of the gel network structure, thus strengthening gel rigidity (G' and tan δ) of WRS/FG mixtures. These transformations may promote the application of hydrocolloid in developing starch-based food products with good quality, especially for improving the stability and texture of host foods. Notably, with increasing applications of hydrocolloids in frozen starchy foods and colon friendly foods, the freeze-thaw stabilities and nutritional properties of starchy foods need to be further focused.

4. Conclusion

The effects of FG addition on the physicochemical properties (thermal and pasting properties, rheological behavior, and digestibility) and aggregate structure (helical structure, short-range ordered structure, crystalline structure, lamellar structure, microstructure) of WRS were systematically elucidated. The thermal stability and pasting viscosity of WRS increased with the FG addition, from 90.40 °C to 92.50 °C (T_c) and 839.00 mPa·s to 1238.00 mPa·s (FV), which is beneficial for preparing baked foods. Also, FG effectively improved the gel rigidity (*i.e.*, increased G' and decreased tan δ) of WRS gels, making starch gels have a better mouthfeel and appearance, which is suitable to produce jelly foods. These results may be attributed to the compact structure formed by hydrogen bonds in the WRS/FG matrix, which was supported by the increased $R_{1047/1024}$ value, *RC*, the density or firmness of the aggregate structure (α) and microphase heterogeneity (ε). Consequently, it is inferred that pasting, gelling, and digestibility can be intentionally controlled by incorporating flaxseed gum (FG). The information provided in this study will help people to better understand the effects on starch functionality modification through hydrocolloids, promoting practical application and increasing the economic effect of starch and gums in food industry. Notably, because of the complexity of the real food systems, future investigations should focus on the regulation effect from more complicated foods or models.

CRediT authorship contribution statement

Xinping Zhang: Writing – review & editing, Writing – original draft, Visualization, Validation, Methodology, Investigation, Formal analysis, Data curation, Conceptualization. **Huishan Shen:** Writing – review & editing, Visualization, Validation, Methodology, Investigation, Formal analysis, Data curation, Conceptualization. **Jingyue Qiao:** Visualization, Validation, Methodology, Formal analysis. **Shuaihao Li:** Visualization, Methodology, Formal analysis. **Xiaojuan Yang:** Visualization, Formal analysis. **Xingli Liu:** Visualization, Formal analysis. **Yanyan Zhang:** Writing – review & editing. **Hua Zhang:** Writing – review & editing, Supervision, Resources, Funding acquisition. **Xuewei Zhao:** Writing – review & editing, Supervision, Resources. **Hongwei Wang:** Writing – review & editing, Validation, Supervision, Resources, Methodology, Funding acquisition. **Fengwei Xie:** Supervision, Resources, Project administration, Methodology, Conceptualization.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

Data will be made available on request.

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Appendix A. Supplementary data

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